

# **Plant Uptake of Organic Pollutants from Soil: A Critical Review of Bioconcentration Estimates Based on Models and Experiments**

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Plant bioconcentration ratio review and evaluation

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## Abstract

The role of terrestrial vegetation in transferring chemicals from soil and air into specific plant tissues (stems, leaves, roots, etc.) is still not well characterized. We provide here a critical review of plant-to-soil bioconcentration ratio (BCR) estimates based on models and experimental data. This review includes the conceptual and theoretical formulations of the bioconcentration ratio, constructing and calibrating empirical and mathematical algorithms to describe this ratio and the experimental data used to quantify BCRs and calibrate the model performance. We first evaluate the theoretical basis for the BCR concept and BCR models and consider how lack of knowledge and data limits reliability and consistency of BCR estimates. We next consider alternate modeling strategies for BCR. A key focus of this evaluation is the relative contributions to overall uncertainty from model uncertainty versus variability in the experimental data used to develop and test the models. As a case study, we consider a single chemical, hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), and focus on variability of bioconcentration measurements obtained from 81 experiments with different plant species, different plant tissues, different experimental conditions, and different methods for reporting concentrations in the soil and plant tissues. We use these observations to evaluate both the magnitude of experimental variability in plant bioconcentration and compare this to model uncertainty. Among these 81 measurements, the variation of the plant/soil BCR has a geometric standard deviation (GSD) of 3.5 and a coefficient of variability (CV-ratio of arithmetic standard deviation to mean) of 1.7. These variations are significant but low relative to model uncertainties--which have an estimated GSD of 10 with a corresponding CV of 14.

**Keywords:** Soil-plant uptake, bioconcentration, transport models, model uncertainty, hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX)

## Introduction

Plant-to-soil and plant-to-air bioconcentration ratios (BCRs) are used to relate chemical concentrations measured in different vegetation tissues to concentrations in the soil supporting that vegetation. In spite of continuing laboratory and field studies, the role of terrestrial vegetation in transferring chemicals from soil and/or air into specific plant tissues (stems, leaves, roots, etc.) is still not well understood. The lack of chemical-, plant-, and site-specific BCR measurements has led to a reliance on empirical and process-based models to explain and predict the fate of chemicals in air/plant/soil systems. The uncertainties associated with both BCR experiments and the models derived from these experiments often limit the reliability of these models for making estimates of chemical uptake and estimating exposure to humans and ecological receptors.

Chemicals in the environment are transferred to plant parts from both air and soil. Efforts to model and evaluate plant uptake of chemicals tend to focus separately on air-plant and soil-plant transfers. The focus of the models and data reviewed in this paper is on the soil-to-plant pathways. We recognize that field and chamber studies show that, in addition to soil uptake, both gas-phase uptake and particle-phase deposition can transfer chemicals from air to vegetation [1-8]. But we restrict our evaluation to the body of theoretical and experimental work that addresses plant-to-soil BCRs. These studies have led to models that explain the relative magnitude of soil-plant transfer based on chemical properties.

Chemicals in soil enter plants primarily through the root system and the degree of uptake from soil into root tissues appears to be proportional to the octanol/water partition coefficient,  $K_{ow}$  [9]. Thus, studies on the bioconcentration of nonionic organic chemicals have focused on correlations between partition factors and chemical properties that express relative solubility, such as  $K_{ow}$ . But the pool of data available for making soil-plant uptake models is limited. As a result, only relatively simple models are available to express plant uptake in terms of chemical properties such as  $K_{ow}$ .

In this paper we provide a critical review of plant-to-soil bioconcentration ratio (BCR) estimates based on models and experimental data. The key focus is on the reliability of BCR estimates and the sources and magnitude of uncertainty associated with BCR estimates in risk assessments and other impact studies. In the sections below, we consider the conceptual and theoretical formulations of the bioconcentration ratio, formulating and calibrating empirical and mathematical algorithms to describe this ratio and the experimental data used to quantify BCRs and calibrate the model performance. We review the uncertainties in conceptual vegetation mass-balance models by evaluating the theoretical basis for these models and considering how lack of conceptual clarity leads to model uncertainty and misinterpretation. We next consider alternate modeling strategies and the associated changes in the reliability, fidelity, and transparency of the models. Here we confront such issues in BCR model development as selecting the general model structure, how many compartments to include, and selecting the outcome metric—total fresh-mass concentration, fugacity, liquid-phase concentration, dry-mass concentration, etc. . Finally, there is the issue of how measurement uncertainty and variability for calibration data define the reliability of BCR values derived from experiments and how this compares to model performance uncertainty. For this we evaluate both the magnitude and primary sources of

measurement variability and uncertainty in plant uptake experiments and compare these uncertainties to conceptual uncertainty and model formulation uncertainty. Here we consider a single widely-studied chemical, hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), and focus on the variability of BCR measurements obtained from a number of experiments with different plant species, different plant tissues, different experimental conditions, and different methods for reporting concentrations in the soil and plant tissues.

### **Plant Bioconcentration Ratios: Concepts, Models, and Measurements**

In this section, we review the plan-soil BCR in terms of conceptual model formulation, mathematical model formulations, and model calibrations with experimental data. Our goal is to review the current literature on plant uptake to provide a critical evaluation of how well the soil-to-plant transfer pathway is understood and quantified. We give particular attention to how gaps in conceptual understanding, limited experimental observation, and key assumptions contribute to the overall uncertainty in BCR estimates derived from experimental observation and models.

Chemicals are transferred from soil to various plant parts through two stages [6, 9]. In the first stage, the chemical can be transferred from soil to the vegetation via one of four pathways—(1) uptake through the roots; (2) volatilization of the chemical from soil with the chemical vapors trapped by the plant leaves; (3) rain-splash, where soil particles are dispersed onto the leaf surfaces following impact of rain drops on the soil surface, and (4) wind induced suspension of soil particles that subsequently fall out onto the surfaces of vegetation. In the second stage the chemical moves from roots and/or leaves to other components of the plant (translocation). These stages are illustrated in **Fig. 1** where various concepts of the BCR are also illustrated.

### *Conceptual Models of Plant Uptake*

In the current scientific literature, plant-soil bioconcentration is used to describe the ratio of the concentration measured in vegetation to a concentration in the soil supporting that vegetation. However, defining “concentration in vegetation” is often difficult, particularly in larger plants where the roots, stems, and leaves all have significantly different contaminant concentrations. A plant-to-soil BCR expresses the observed or predicted ratio of contaminant concentration in plant parts relative to concentration in soil. There are at least five ways to define a plant BCR relative to chemical concentration in the soil solids (dry mass) or relative to the concentration in the water solution in soil. These definitions are summarized in **Table 1**. The choice of definition impacts both the interpretation of BCR measurements and the form of the model used to express them.

### *General Model Formulations for the Plant-to-Soil BCR*

In order to evaluate the performance of BCR models, we consider here a range of existing models that express plant uptake in terms of the chemical properties such as  $K_{ow}$  and quantitative structural factors such as the molecular connectivity index (MCI). In the following paragraphs we describe the principal modeling options proposed in the current literature. In the next subsection we discuss tools for determining the reliability of these model for predicting plant uptake from soil.

An early effort by Briggs et al. [10-11] used  $K_{ow}$  to estimate uptake by roots, transpiration stream, and stems from soil solution. Others have since developed alternative models building on the Briggs et al. approach. In particular, Travis and Arms [12] who reviewed measurements of bioconcentration reported in the literature for 29 persistent organo-chlorines in plants to develop



a  $K_{ow}$ -based empirical regression model of plant-soil BCR (on a dry-mass basis) in above-ground plant parts. Dowdy and McKone [13] compared the precision and accuracy of the MCI relative to  $K_{ow}$  as a predictor of bioconcentration from the soil matrix into above- or below-ground vegetation tissues. However, Riederer [14], McKone [15], and Dowdy and McKone [13] have all reported the low reliability of using simple plant/soil partition coefficients to characterize uptake. McFarlane et al. [16] have demonstrated experimentally that chemicals with similar values of  $K_{ow}$  can have drastically different long-term partitioning.

Based on experiments with 18 radio-labeled *O*-methylcarbamoyloximes and substituted phenylureas, Briggs et al. [10] measured partitioning between barley roots and nutrient solutions. They refer to this partition coefficient as the root concentration factor, RCF, which is defined in Table 1, and proposed estimation equations based on these data. Other researchers have applied different data sets to propose alternative formulations for the RCF regression with  $K_{ow}$  [17, 18]. In order to represent the translocation of chemicals from the root-soil pore water to the stem of plants, Briggs et al. [10] defined the transpiration stream concentration factor (TSCF) and formulated a regression model for this BCR based on the same experimental set used for RCF. The TSCF accounts for the reduction in concentration in the pore water as it crosses the root membrane and moves through the xylem to the stem. Thus, the TSCF represents the ratio of contaminant concentration in the xylem stream of the stem, mg/L, to contaminant concentration in soil solution, mg/L. Hsu et al. [19] and Burken and Schnoor [18] have proposed alternate constants for the Briggs et al. TSCF model using alternate experimental data sets. Briggs et al. [11] also made measurements of chemical uptake in macerated stems from solutions for 8 *O*-methylcarbamoyloximes and substituted phenylureas in barley shoots to develop a model of the stem to soil-solution concentration ratio (SCF).

Trapp and Matthies [20] developed a one-compartment differential mass balance model for air-to-plant and soil-to-plant uptake. Included in this model are uptake from soil through the transpiration stream, gaseous deposition, volatilization from leaves, chemical transformation, and growth dilution. Their model provides a method for assessing a transient and long-term BCR from soil solution to leaves. Hung and Mackay [21] followed a similar approach to mass balance as Trapp and Matthies [20], but used three compartments—roots, stem, and leaves—instead of one compartment. The Hung and Mackay model [21] provides a similar opportunity for constructing a transient and plant-specific method for calculating BCFs, but requires many more plant- and chemical-specific parameters.

Dowdy and McKone [13] used the MCI as a quantitative structure-activity relationship (QSAR) for predicting plant-soil bioconcentration. They report that the normal path first-order MCI could reliably predict plant biotransfer for nonpolar compounds without any need for correction factors. But they found that extension of this predictive model to estimate biotransfer for polar compounds requires an adjustment of the index with correction factors to account for polar group effects.

Chiou et al. [22] revisited the data of Briggs et al. [10], Trapp et al. [23] and earlier data for pesticides to propose a novel modeling approach based on the following assumptions: (1) overall transport from soil is driven by soil-water concentration; (2) each volume element within the plant attains equilibrium with the sap in that component; and (3) the contaminant concentration in each volume element can be scaled to the soil-water solution.

*Model Residual Error, Model Differences, and Model Uncertainty*

In order to compare different modeling approaches in terms of model error, we consider how well each model matches its own calibration set, how well the models compare with each other and how well each model tracks an ensemble set of data, pulled together from different studies.

In order to evaluate how well a model matches its own calibration set, we use the standard error of the estimator (SEE). The SEE for a BCR derived from any estimation scheme (i.e., process based or regression model) is calculated according to the following formula [15, 24].

$$SEE \text{ of } \log B_x = \sqrt{\frac{1}{(n-2)} \sum_{i=1}^n [\log B_x^{msd} - \log B_x^{est}]^2} \quad (1)$$

where  $B_x$  refers to the estimated BCR,  $n$  is the number of chemicals used in the estimation protocol,  $B_x^{msd}$  refers to the measured BCR, and  $B_x^{est}$  refers to the corresponding estimated BCR value. We also compare results by considering the geometric standard deviation (GSD) among a set of observations and the coefficient of variation (CV) for a set of observations, the ratio of arithmetic standard deviation to the arithmetic mean,

$$CV = \text{Stdev}/\mu. \quad (2)$$

When we have the GSD and geometric mean (GM) from a set of observations or from the residual error of a model calibration exercise and need to estimate CV, we use Monte Carlo sampling from a lognormal distribution with the specified GSD and GM to estimate the arithmetic mean ( $\mu$ ), Stdev, and CV.

*Variations in Mathematical Formulations for Models of Plant Uptake from Soil*

In the sections below we compare key features of a representative set of model formulations that we used to explore variations in BCR estimates among the models.

We first consider RCF. Below is the Briggs et al. [10] estimation equation for RCF from their experiments with 18 radio-labeled *O*-methylcarbamoyloximes and substituted phenylureas taken up by barley roots from nutrient solution:

$$\log (\text{RCF} - 0.82) = 0.77 \log K_{ow} - 1.52 \pm 0.12 \quad (n=18, R^2=0.97) \quad (3)$$

The term  $\pm 0.12$  is the SEE for the log RCF. Based on studies of 9 carbon-14-labeled organic chemicals in barley and cress, Topp et al. [17] developed a second version of this regression model:

$$\log \text{RCF} = 0.630 \log K_{ow} - 0.959 \quad (n=9, R^2=0.80) \quad (4)$$

Burken and Schnoor [18] used 11 carbon-14-labeled compounds taken up by poplar-tree shoots to obtain a third version of this model:

$$\log (\text{RCF} - 3.00) = 0.65 \log K_{ow} - 1.57 \quad (n=11) \quad (5)$$

Equations 3, 4, and 5 give similar results for RCF at  $\log K_{ow} = 2$ , but at  $\log K_{ow} = 0$  their RCF estimates differ by almost a factor of 30 and at  $\log K_{ow} = 6$  they differ by a factor 5. This is a much larger variation than the residual error reported in Equation 3, which we assume to be roughly the same for all three models.

Briggs et al. [10] formulated the following regression model for TCSF based on their 18 experiments with barley roots and nutrient solutions,

$$\log \text{TSCF} = 0.784 \times \exp \left[ -\frac{(\log K_{ow} - 1.78)^2}{2.44} \right] \pm 0.27 \quad (n=18, R^2=0.61) \quad (6)$$

Hsu et al. [19] developed an equation of similar form using soybean roots, but with slightly different regression parameters:

$$\log \text{TSCF} = 0.70 \times \exp \left[ -\frac{(\log K_{ow} - 3.07)^2}{2.78} \right] \quad (7)$$

Burken and Schnoor [18] used carbon-14-labeled compounds taken up in poplar-tree shoots to obtain a third version of this model:

$$\log \text{TSCF} = 0.756 \times \exp \left[ -\frac{(\log K_{ow} - 2.50)^2}{2.58} \right] \quad (n=11) \quad (8)$$

When  $\log K_{ow}$  is in the range 0 to 5, the estimate of TSCF differs among Equations 6 through 8 by an order of magnitude. We note that this difference is much greater than the residual error shown in Equation 6.

In order to obtain an estimate of the SCF, Briggs et al. [11] made measurements of chemical uptake in macerated stems from solutions for *O*-methylcarbamoyloximes and substituted phenylureas in barley shoots and obtained the following expression for the stem to solution concentration:

$$\log K(\text{stem/solution}) = 0.95 \log K_{ow} - 2.05 \quad (n=8, R^2=0.96) \quad (9)$$

Under the assumption that the contribution of the aqueous phase in stems to the uptake of chemical by stems is similar to that in roots (that is 0.82), Briggs et al. [11] used Equation 7 to develop the following relationship for the stem/xylem-sap partition coefficient:

$$\log [K(\text{stem/xylem sap}) + 0.82] = 0.95 \log K_{ow} - 2.05 \quad (10)$$

From Equation 10, Briggs et al. [11] developed the stem concentration factor (SCF). Based on the assumption that SCF is the product of  $K(\text{stem/xylem sap})$  and TSCF (from Equation 4) they obtained the following model formulation of SCF,

$$SCF = (0.082 + 0.0089 \times K_{ow}^{0.95}) \times \left\{ 0.784 \times \exp \left[ -\frac{(\log K_{ow} - 1.78)^2}{2.44} \right] \right\} \quad (11)$$

The widely used Travis and Arms [12] correlation of plant-soil bioconcentration (on a dry-mass basis) in aboveground plant parts with  $K_{ow}$  is expressed as

$$\log BCR_{d,ss} = 1.58 - 0.58 \log K_{ow} \pm 0.73 \quad (n=29, R^2=0.525) \quad (12)$$

The error term in Equation 12,  $\pm 0.73$ , was not provided by Travis and Arms [12] but was calculated by McKone [15] using the SEE.

The Trapp and Matthies [20] model provides a method for assessing a transient and long-term BCR from soil solution to leaves. When the concentration of contaminant in air is low relative to plant, their model reduces to the following form for expressing the transient and plant-specific biotransfer factor,

$$BCR_{f,sw}(t) = C_L(t)/C_w = \frac{TSCF \times Q}{m_L a} \times [1 - \exp(-at)] \quad (13)$$

where

$C_L$  = concentration in plant leaves, mg/kg;

$C_w$  = concentration of contaminant in soil solution, mg/L;

$a = Ag/(K_{LA}V_L) + k_e + k_g$

$A$  = leaf area,  $m^2$ ;

$g$  = air-to-leaf conductance,  $m/d$ ;

$K_{LA}$  = leaf-air partition coefficient,  $mg/m^3(\text{leaf})$  per  $mg/m^3(\text{air})$ ;

$V_L$  = volume of leaves,  $m^3$ ;

$k_e$  = rate constant for elimination by chemical transformation (i.e. metabolism) within the leaves,  $1/d$ ;

$k_g$  = growth dilution rate constant,  $1/d$ ;

TSCF = transpiration stream concentration factor;  $L(\text{soil solution})/L(\text{transpiration stream})$ ;

$Q$  = transpiration flow from soil through stems,  $L/d$ ; and

$m_L$  = fresh mass of the leaves,  $kg$ .

As time,  $t$ , become large with respect to  $a$  and for  $\log K_{ow}$  less than 4, Equation 13 tends to track the SCF proposed by Briggs et al. [11] (Equation 11). When  $\log K_{ow}$  is above 4, this BCR tends to be significantly lower than SCF. However, this is expected since this expression provides a mass-balance for leaves, whereas the SCF is for stems.

Using the polar-corrected MCI,  $^1\chi_{pc}$ , Dowdy and McKone [13] developed the formulations below for bioconcentration ratios (BCRs) for roots relative to soil (root-s) and above ground plant relative to soil (abg-s).

$$\log BCR_{\text{root-s}} = 0.718(^1\chi_{pc}) - 2.372 \pm 0.90 \quad (n=16, R^2=0.84), \quad (14)$$

$$\log BCR_{\text{abg-s}} = -0.204(^1\chi_{pc}) + 0.589 \pm 0.34 \quad (n=30, R^2=0.83) \quad (15)$$

The units of  $BCR_{root-s}$  are L(soil solution)/kg (plant fresh mass) and the units of  $BCR_{root-s}$  are kg(soil dry mass)/kg(plant dry mass). Both terms have large residual errors because of the large range of physical-chemical properties of the organic chemicals addressed in this regression.

Chiou et al. [22] expressed the concentration of a contaminant in either the whole plant or a specific part of the plant (i.e., roots, stems, leaves, etc.) ( $C_{pt}$ ) on a wet-mass basis relative to soil-water concentration  $C_w$  using the following expression:

$$C_{pt} = \alpha_{pt} C_w [\phi_{pom} K_{pom} + \phi_{pw}] \quad (16)$$

The term  $\alpha_{pt}$  is called “the quasi-equilibrium factor”, which describes the approach to equilibrium of any absorbed contaminant in the plant (or in a part of it) with respect to the same contaminant in soil solution.  $K_{pom}$  is the contaminant partition coefficient between plant organic matter and water,  $\phi_{pom}$  is the total weight fraction of the organic matter in the plant, and  $\phi_{pw}$  is the weight fraction of water in the plant, either for the whole plant or for a specific part of it. In this model  $\alpha_{pt}$  may be viewed as the ratio of the respective concentrations in plant water and external water. Thus,  $\alpha_{pt} = 1$  denotes the state of equilibrium. The magnitude of  $\alpha_{pt} (\leq 1)$  is a measure of the extent of approach to equilibrium. If passive transport is the dominant uptake process,  $\alpha_{pt}$  should not exceed 1, except for highly unusual situations; if the uptake involves an active process,  $\alpha_{pt}$  may however exceed 1. Chiou et al. [22] determined values of  $\alpha_{pt}$  based on the data of Briggs et al. [10], Trapp et al. [23], and earlier data for pesticides and found that alpha varied roughly within an order of magnitude among the various experimental results.



### *New Opportunities for Model Calibration and Testing*

In addition to the published experimental data that have been used to develop current bioconcentration models [10, 11, 12, 16, 17, 19, 20], there have been a number of recent experimental assessments that provide additional opportunities to evaluate the performance of plant-to-soil bioconcentration models. For example, Sicbaldi et al. [25] have compiled new data on root uptake of pesticides. Kraaij and Connell [26] have measured the root-to-soil bioconcentration of a series of chlorobenzenes in soybean roots. Kipopoulou et al. [27] have measured the plant-to-air and/or plant-to-soil bioconcentration of polycyclic aromatic hydrocarbons in vegetables grown in an industrial area. The US EPA [28] have compiled data on soil and plant concentration data that provides a potential opportunity to assess plant-to-soil partition factors for a number of dioxin congeners.

### *Compilation of Experimental Data Showing BCR Variability for Single Chemical*

We focus on the collection of BCR data for a single chemical in order to evaluate how experimental variability impacts BCR estimates,. In the next section where overall BCR reliability and uncertainty are considered, we compare this experimental variability to both to conceptual model uncertainty and to model formulation uncertainty. As the single chemical of interest, we select hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), a compound that has been studied extensively. We assembled a number of reports and journal papers describing experiments in which plants were grown in soil or water containing a measured concentration of RDX and in which the plant tissues were sampled for RDX. Through this process we obtained 81 different experimental observations of plant-to-soil BCRs for RDX from experiments with different plant species, plant tissues, experimental conditions, and methods for reporting

concentrations in the soil and plant tissues. These measurements are summarized in **Table 2**. The cited studies include experimental data on ten different plant types--alfalfa, beans, corn, lettuce, tomato, carrot, cucumber, nutsedge, radish, and spinach; seven different plant tissues--roots, stem, leaves, pods, fruits, tassels, and seeds; both dry- and fresh-mass based measures of plant concentrations; measurements of chemical concentration and of radio-labeled tracers; and both dry- and solution-based measures of soil concentrations.

The results in Table 2 include observations from six different reports that provide results from multiple experiments, and different experimental protocols. Cataldo et al. [29] used radio-labeled RDX to measure uptake from contaminated soil into the roots, stems, leaves, pods, and seeds of bean plants. They also used beans grown in hydroponic solutions with radio-labeled RDX to compare with their field results. Cataldo et al. [29] report all plant concentrations on a fresh-mass basis. But they report their soil concentrations on a dry mass basis and also report soil organic carbon. Checkai and Simini [30] used hydroponic studies to measure concentrations of RDX on a fresh-mass and dry-mass basis in tissues of plants grown in a growth solution with a specified RDX concentration. They carried out these experiments for bush bean fruit, corn stover, lettuce leaves, alfalfa shoot, radish roots, and tomato fruit. Price et al. [31] reported the dry mass plant tissue concentration relative to soil fresh-mass concentration for experiments that exposed corn kernel, corn stover, lettuce leaves, nutsedge, and tomato fruit, to RDX in soil and soil solution. Harvey et al. [32] used radio-labeled RDX in 1- and 7-day hydroponic studies to measure uptake in bean roots, bean stems, and bean leaves. Fellows et al. [33] reported the dry mass plant tissue concentration relative to soil dry-mass concentration for corn ear, corn stem, corn leaves, corn tassel, alfalfa roots, alfalfa shoot, alfalfa seeds, carrot shoot, spinach root,

spinach shoot, and spinach seeds. Larson et al. [34] measure the uptake on a dry mass basis in the leaves and tassels of corn grown in RDX contaminated solution.

The compilation of BCR values in Table 2 shows large variations in the experimental conditions and interpretations of measurements. In order to harmonize these data we converted all the results to an equivalent BCR with units of concentration in plant fresh mass divided by soil solution concentration, that is  $\mu\text{g/kg}$  (fresh plant mass) per  $\mu\text{g/L}$  (soil water). For some experiments this required the conversion of a reported soil dry mass concentration to an equivalent soil solution concentration for which we used the follow conversion:

$$C(\text{soil fresh mass}) = C(\text{plant dry mass})/K_d = C(\text{plant dry mass})/(K_{oc}f_{oc}) \quad (17)$$

Where  $K_d$  is the soil-water distribution coefficient ( $\mu\text{g/kg}[\text{soil}]$  per  $\mu\text{g/L}[\text{water}]$ ), which is estimated as the product of the organic carbon partition factor  $K_{oc}$  ( $\mu\text{g/kg}[\text{organic carbon}]$  per  $\mu\text{g/L}[\text{water}]$ ) and the fraction of organic carbon in soil,  $f_{oc}$ . For RDX, we used a  $K_{oc}$  value equal to 100 [35]. The values we used for  $f_{oc}$  are reported in Table 2 and obtained from the cited references. For some experiments we had to convert a reported plant dry- mass concentration ( $\mu\text{g/kg}$ ) to an equivalent plant fresh-mass concentration ( $\mu\text{g/kg}$ ). For this we used the following conversion:

$$C(\text{plant fresh mass}) = C(\text{plant dry mass}) * f_{\text{dry-mass}} \quad (18)$$

Where  $f_{\text{dry-mass}}$  is the fraction of plant fresh mass that will become dry mass under experimental conditions. In Table 2, we list the value used for this parameter when it is required. These values were either obtained from the cited reference or assigned a default value of 0.2 when there is no reported value.

We applied a statistical analysis to the values of the RDX BCR in Table 2 to determine the characteristics of the range and spread of these results. The arithmetic mean and standard deviation of the 81 RDX BCR values given in Table 2 are, respectively, 2.3 and 4.0, corresponding to a CV of 1.7. The GM and GSD are respectively 1.1 and 3.5.

### **The Reliability of Plant Bioconcentration Estimates**

In order to evaluate the reliability of the models and experimental data currently available to assess plant-soil BCR, we consider three issues. First we consider uncertainties that arise from conceptual model formulation. These results are primarily qualitative and subjective, but set the stage for the more quantitative reliability questions. Next, we consider the likely variation in results from mathematical models used to characterize plant uptake from soil. Our focus here is on the residual error associated with the use of models to estimate the BCF for an unspecified soil/plant/plant tissue system. Finally, we consider the estimation error associated with using our collection of experimental observations for RDX to make an estimate for the BCF for RDX in an unspecified soil/plant/plant tissue system and compare this to the uncertainty associated with making BCR estimates from models.

#### *Uncertainties in Model Formulation*

Based on our review of available plant biotransfer models, we find that the range of conceptual model formulation options introduces significant, but difficult to quantify, uncertainty to the process of modeling BCRs. A key source of uncertainty and potential error is in the definition of bioconcentration and selection of the appropriate soil concentration metric (dry mass, fresh mass, soil solution, etc.) and the representative plant concentration metric, plant species, and plant tissue. But even when the concept and metric of BCR is clear, there are

additional key uncertainties introduced by the options and limitations for measurements that support the conceptual model. As observed previously by Paterson et al. [9], three types of factors affect the uptake and distribution of chemical compounds within plants. First and foremost are physicochemical properties of the compound such as water solubility, vapor pressure, molecular weight, and octanol/water partition coefficient. Second are environmental characteristics including temperature of the soil as well as organic and mineral matter content and water content of the soil. Third are plant characteristics such as the type of root system, shape and composition of the leaves, and lipid (oil) content. These three issues limit the options for reducing uncertainty at the model formulation stage. The reliability of any model based on physicochemical properties is also limited by the accuracy and availability of these properties. Physicochemical properties for many compounds are not accurately known. Soil characteristics are highly variable from one location to another and also vary seasonally at a single location. In addition, plant characteristics also vary significantly from one plant species to another, and seasonally in the same plant.

#### *BCR Model Uncertainty due to Residual Calibration Error and Model Differences*

Our confidence about the ability of models to provide estimates of BCRs derives both from the ability of a single model to match its calibration set of chemicals and from the consistency with which different models produce similar BCR values.

Our evaluation reveals that the residual errors (SEE) of the log BCR among different models relative to measured values range between 0.12 to 0.9 log units, which is equivalent to a geometric standard deviation (GSD) range of 1.3 to 8, corresponding to a coefficient of variation

(CV) of between 0.27 and 8.6. While this is not a measure of overall model uncertainty, it does measure how well a model fits its own calibration set.

The more important issue, however, is the expected reliability of the models for predicting BCR values for chemicals not included in their calibration set. This test has rarely been applied, but we can use the results above to make some general comparisons. In **Fig. 2** we provide a plot of the BCR estimates for several models with  $\log K_{ow}$  as the predictive parameter. We also include in this figure the calibration data for each model to get a sense of how the results converge or diverge among the different models. To construct this figure, we converted all the measured values and the model predictions to equivalent plant-fresh-mass-soil-solution BCR ratios. This figure has a log-log scale and reveals a significant divergence among the models. For example, we see here that both the TSCF and the SCF models of Briggs et al [10, 11] predict a rapid fall in BCR above a  $K_{ow}$  of 3 for the TSCF models and above a  $K_{ow}$  of 6 for the SCF model. But the Briggs et al. [10,11]-type models for RCF, TSCF, and SCF have not been tested for compounds with  $\log K_{ow}$  above 5 to confirm this rapid decrease. Moreover, this is not consistent with experimental observations of BCR for some higher  $K_{ow}$  compounds. Nevertheless, the Briggs et al. [10,11]-type models appear to have relatively high reliability for fitting observations for chemicals with  $0 < \log K_{ow} < 5$  or 6. Although the Travis and Arms [12] BCR model tends to track along the center range of the measured values shown in Fig. 2, we note that it should only be used for compounds with  $1 < \log K_{ow} < 9$ , i.e., the range in which the model was calibrated.

In Fig. 2 we include two (dark) lines that capture the roughly 4 standard deviation interval (95% confidence interval) around both the experimental and model results. These lines indicate

that the differences among both the models and the observations used to construct them have an estimated GSD of 10 with a corresponding CV of 14. These are large uncertainties. But, this ensemble uncertainty is not significantly higher than the residual estimation error of models that are designed to capture variability of BCR across a broad range of chemical properties. For example the residual error of the MCI-based regression model [13], as reflected in Equation 14 has GSD of 8.

*BCR Uncertainty for a Single Chemical due to Experimental Variability*

In order to provide an example of the uncertainty due to variability, we developed a cumulative probability plot for the 81 observed values of RDX BCR reported in Table 2. We developed this probability plot based on their rank order using the relationship:

$$\text{Cumulative Probability} = (\text{rank}-0.5)/81 \quad (19)$$

The resulting cumulative probability plot for the 81 RDX BCR observations is shown in **Fig. 3** as a series of open circles. Also plotted for comparison in Fig. 3 is a cumulative lognormal probability distribution with GM 1.1 and GSD 3.5. We see here that the lognormal distribution with these moments provides a good fit to the variance in BCR from the set of experimental observations.

We focus on the single compound RDX to gain insight on the relative contributions of model uncertainty and experimental variability to our ability to estimate a BCR for a selected organic chemical. **Fig. 4** summarizes our comparison presented as two cumulative probability distributions. The two lognormal distributions in Fig. 4 show the relative variance attributable to experimental variability and to model uncertainty for estimating the BCR value for a chemical such as RDX that has an expected BCR value of 1.1. The lognormal distribution for variance in

BCR obtained from 81 experiments is taken from Fig. 3 (GSD of 3.5; CV of 1.7). We estimate that, in the absence of any chemical and site-specific experimental information, the expected uncertainty of a BCR model for RDX can be represented by a log normal distribution with a GM of 1.1, a GSD of 10, and a CV of 14. This is the second cumulative distribution shown in this figure. Also shown are estimated 95% confidence intervals on the BCR estimate based on model uncertainty and experimental variability.

In order to compare the relative contribution of experimental variability and model uncertainty, we used the  $R^2$  squared statistic, which is a measure of variance in one set of observations explained by a second set. We computed  $R^2$  from the two the sources of variation--the variance of the experimental measures about the mean BCR and the variance attributable to the uncertainty in results from the model-based estimates of the BCR. We did this by using random sampling to obtain 3000 samples from the lognormal distribution of model uncertainty and 3000 samples from the lognormal distribution for experimental variability. Based on the correlation coefficient  $R$  between these two samples, we obtained an  $R^2$  equal to 0.57. This indicates that a significant fraction of the overall BCR estimation uncertainty is attributable to experimental variability.

## **Discussion and Findings**

Our critical review of the range and reliability of methods for estimating plant-soil BCR offers insight for environmental scientists who must interpret and apply plant-uptake estimates obtained from models and experiments. In this assessment we have emphasized the importance of confronting uncertainties at each stage of the model development and application. We see that uncertainty emerges at the conceptual model stage as well as during mathematical model



formulation and calibration and in model applications. The results above show that uncertainty is not simply a variance propagation or “Monte Carlo” assessment that is used to propagate parameter variance at the model application stage. Instead it is a process that begins at the earliest stage of model development and accrues through model formulation and specific applications.

In the studies reviewed in this paper, we find that important uncertainties arise at the first stage of model development—the concept formulation. For plant uptake models that address competing soil-root-leaf and soil-air-leaf pathways, conceptual uncertainties remain a dominant source of overall uncertainty. An important contributor to this conceptual uncertainty is the lack of a consistent definition of BCR for soil uptake in both experiments and models. This leads to confusion and inconsistency in the use of BCR. Because this type of uncertainty is difficult, if not impossible, to quantify, we must develop qualitative methods and classifications to communicate this important source of uncertainty.

In evaluating model formulation, we observe large differences among models in their predictions of BCR, but we discovered no clear basis for selecting one model as more accurate than another. The residual errors reported for many of the models in fitting their calibration data leads us to believe that we do not yet have sufficient data to formulate accurate models. But our review of experiments for the single chemical RDX reveals that much of the uncertainty is attributable to the lack of precision and variability of experimental data used to obtain the BCR values used to calibrate models. It appears that there are advantages to using more than one compartment in formulating BCR models. But lack of experimental data and the poor state of conceptual knowledge suggests that model uncertainty cannot be reduced by adding large numbers of compartments to the models. Too many plant components in our models lead to over

specification. But a single compartment model can miss the combined effect of root and shoot uptake processes.

The interaction among measurements, conceptual models, and models leads to the conclusion, contrary to our initial expectation, that it may not be possible to distinguish the relative contributions of overall uncertainty from conceptual uncertainty, measurement variability, and model uncertainty. For example, the variations in the value of BCR obtained from the 81 experiments we considered for RDX span a rather wide range. But it is not clear how much of this variation is attributable to experimental uncertainty and how much to inadequate conceptual models. Often the conceptual model is used to design experiments so that an incorrect conceptual model leads to measurements that are difficult to interpret when they are inconsistent with the concept. Perhaps the variance in the experimental values would be much lower if we understood better how BCR is affected by variables whose impact is not yet fully understood—for example temperature, soil properties, etc. Similarly if we really had a complete and thorough conceptual understanding of the process of uptake, then choosing a mathematical equation would likely be less uncertain. That is, the mathematical model formulation may only appear uncertain because we are using mostly-empirical mathematical relationships to describe a process that we do not understand well enough at a conceptual level. So it is not clear whether we classify this as uncertainty in the mathematical model formulation, or as uncertainty in the conceptual model.

In applying model performance evaluation to plant uptake modeling, the results and discussion above lead us to a number of key findings. These include:

The conceptual formulation of the bioconcentration ratio has an important, but at this point difficult to quantify, contribution to overall uncertainty. In particular, the concept of different

plant components, the selection of dry- versus fresh-mass concentrations, and the use of dynamic or steady state concentration ratio strongly impact the reliability and uncertainty of the resulting BCR model.

When we consider both the performance of models with respect to their calibration experiments and also compare different models, we find that quantitative results for any randomly selected organic chemical have very large model uncertainties. We estimate that in the absence of specific experimental information, the expected uncertainty of a BCR model can be represented by a log normal distribution with a GSD of 10 ( $CV = 14$ ). This means that without additional information on plant species or without plant- and site- specific measurements, we can only expect a model to predict a BCR within  $\pm 1$  log units such that there is a 66% likelihood that the actual BCR value is 10 times higher or lower than the value obtained from a model.

Based on consideration of a large number of experiments for a single, well-studied compound, RDX, we find that experimental measurements of BCR have large experimental variability and that this experimental variability can be represented by a lognormal distribution with a GSD of 3.5 ( $CV = 1.7$ ). This indicates much of our observed model uncertainty most likely derives from experimental variability. This leads to the observation that controlled measurements cannot necessarily remove the large uncertainties that derive from BCR models.

Comparison for RDX of the relative contributions of model uncertainty and experimental variability to uncertainty in BCR estimates indicates that a large fraction of model uncertainty can be attributed to experimental variability.

The variability and complexity of the uptake and transport of chemicals in vegetation cannot be captured by a point-value for BCF, but requires the use of ranges and confidence intervals to communicate the large uncertainties associated with estimating BCRs. In any plant-uptake model used to estimate a BCR, we must develop a process for communicating both the magnitude of the result and the confidence that can be placed in this number. On the part of the assessor this requires a presentation of both qualitative and quantitative uncertainties.

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Table 1. Comparison of various definitions of the plant-soil bioconcentration ratio (BCR).

Description	Formula
BCR for plant dry mass relative to dry soil (soil solids) concentration	$BCR_{d,ss} = \frac{\text{Concentration in dry plant tissue (mg/kg)}}{\text{Concentration in dry soil (mg/kg)}}$
BCR for root (fresh mass) concentration relative to chemical concentration in the water solution in soil (or soil solution) and referred to as the root-concentration factor (RCF)	$RCF = \frac{\text{Concentration in fresh plant roots (mg/kg)}}{\text{Concentration in soil solution (mg/L)}}$
BCR for transpiration stream concentration relative to chemical concentration in the water solution in soil and referred to as the transpiration-stream-concentration factor (TSCF)	$TSCF = \frac{\text{Concentration in transpiration stream (mg/L)}}{\text{Concentration in soil solution (mg/L)}}$
BCR for plant fresh mass relative to chemical concentration the water solution in soil (for example SCF)	$BCF_{f,sw} = \frac{\text{Concentration in fresh plant tissue (mg/kg)}}{\text{Concentration in soil solution (mg/kg)}}$
BCR for plant fresh mass relative to chemical concentration to dry soil (soil solids) concentration	$K_{ps} = \frac{\text{Concentration in fresh plant tissue (mg/kg)}}{\text{Concentration in dry soil (mg/kg)}}$

Table 2. Compilation of bioconcentration ratio (BCR) values for RDX and summarized by plant type/part, supply medium, supply medium concentration, plant concentration and by whether the soil and plant concentrations are on a dry mass (dm) or fresh-mass (fm) basis.

Plant type and part	Supply Medium	Medium Conc. ppb RDX	Cited as dm or fm	Soil $f_{oc}$	Plant Conc. ppb RDX	Cited as dm or fm	Plant dm fraction	BCR result	Ref.
Bean pods	soil	10,000	dm	0.005	1,292	fm		0.06	[29]
Bush bean fruit	water	100	fm		7	fm		0.07	[30]
Bean roots	soil	10,000	dm	0.005	1,500	fm		0.08	[29]
Tomato fruit	soil	50,300	fm	0.030	74,519	dm	0.07	0.10	[31]
Corn kernal	soil	50,300	fm	0.030	6,140	dm	0.34	0.11	[31]
Radish roots	water	100	fm		14	fm		0.14	[30]
Tomato fruit	water	100	fm		16	fm		0.16	[30]
Bean stems	soil	10,000	dm	0.005	3,738	fm		0.19	[29]
Bush bean fruit	water	20	fm		4	fm		0.20	[30]
Bean pods	soil	10,000	dm	0.017	1,502	fm		0.26	[29]
Corn Stover	water	100	fm		126	dm	0.23	0.29	[30]
Lettuce Leaves	water	134	fm		1,600	dm	0.03	0.30	[31]
Radish roots	water	20	fm		6	fm		0.30	[30]
Tomato fruit	water	20	fm		6	fm		0.30	[30]
Tomato fruit	soil	1,770	fm	0.030	7,867	dm	0.07	0.30	[31]
Bean roots	soil	10,000	dm	0.017	1,965	fm		0.33	[29]
Tomato fruit	soil	1,640	fm	0.030	9,719	dm	0.07	0.40	[31]
Corn Stover	water	406	fm		740	dm	0.23	0.42	[31]
Bean roots	soil	10,000	dm	0.072	650	fm		0.47	[29]
Nutsedge	water	406	fm		980	dm	0.20	0.49	[31]
Corn Stover	water	812	fm		1,780	dm	0.23	0.50	[31]
Corn Stover	water	2	fm		5	dm	0.23	0.57	[30]
Bean roots	water	10,000	fm		6,000	fm		0.60	[32]
Bean stems	soil	10,000	dm	0.017	3,924	fm		0.67	[29]
Tomato fruit	soil	7,675	fm	0.030	79,593	dm	0.07	0.70	[31]
Nutsedge	water	812	fm		2,960	dm	0.20	0.74	[31]
Lettuce Leaves	soil	1,770	fm	0.030	10,000	dm	0.05	0.75	[31]
Corn ear	soil	15,000	dm	0.005	112,000	dm	0.20	0.75	[33]
Lettuce Leaves	water	100	fm		77	fm		0.77	[30]
Bean pods	soil	10,000	dm	0.072	1,073	fm		0.77	[29]
Lettuce Leaves	soil	1,640	fm	0.030	9,620	dm	0.05	0.77	[31]
Bean seeds	soil	10,000	dm	0.005	17,475	fm		0.87	[29]
Lettuce Leaves	water	20	fm		18	fm		0.90	[30]
Bean roots	water	10,000	fm		9,000	fm		0.90	[32]
Bean leaves	soil	10,000	dm	0.072	1,346	fm		0.97	[29]
Bean stems	water	10,000	fm		11,000	fm		1.10	[32]
Bean stems	water	10,000	fm		11,000	fm		1.10	[32]
Bean stems	soil	10,000	dm	0.072	1,555	fm		1.12	[29]

Nutsedge	water	134	fm		1,600	dm	0.10	1.22	[31]
Corn Stover	water	20	fm		107	dm	0.23	1.23	[30]
Bean roots	water	5,000	fm		6,200	fm		1.24	[29]
Alfalfa shoot	water	20	fm		25	fm		1.25	[30]
Alfalfa shoot	water	100	fm		131	fm		1.31	[30]
Lettuce Leaves	water	812	fm		21,320	dm	0.05	1.31	[31]
Corn Stover	water	134	fm		1,600	dm	0.11	1.37	[31]
Lettuce Leaves	water	406	fm		11,140	dm	0.05	1.37	[31]
Corn stem	soil	15,000	dm	0.005	212,000	dm	0.20	1.41	[33]
Lettuce Leaves	soil	673	fm	0.030	7,900	dm	0.05	1.55	[31]
Bean roots	water	5,000	fm		9,400	fm		1.88	[29]
Bean leaves	water	10,000	fm		19,000	fm		1.90	[32]
Bean leaves	soil	10,000	dm	0.017	11,937	fm		2.03	[29]
Alfalfa roots	soil	15,000	dm	0.005	313,000	dm	0.20	2.09	[33]
Corn tassel	soil	15,000	dm	0.005	326,000	dm	0.20	2.17	[33]
Bean stems	water	5,000	fm		10,900	fm		2.18	[29]
Bean stems	water	5,000	fm		11,000	fm		2.20	[29]
Bean leaves	soil	10,000	dm	0.005	45,515	fm		2.28	[29]
Bean seeds	soil	10,000	dm	0.017	13,535	fm		2.30	[29]
Lettuce Leaves	soil	5,800	fm	0.051	62,480	dm	0.05	2.39	[31]
Corn tassel	water	1,000	fm		16,000	dm	0.15	2.45	[34]
Alfalfa seeds	soil	15,000	dm	0.005	377,000	dm	0.20	2.51	[33]
Lettuce Leaves	soil	5,800	fm	0.054	62,740	dm	0.05	2.53	[31]
Lettuce Leaves	soil	7,675	fm	0.030	154,000	dm	0.05	2.65	[31]
Alfalfa shoot	soil	15,000	dm	0.005	429,000	dm	0.20	2.86	[33]
Bean seeds	soil	10,000	dm	0.072	4,025	fm		2.90	[29]
Lettuce Leaves	soil	50,300	fm	0.030	1,172,000	dm	0.05	3.07	[31]
Carrot shoot	soil	15,001	dm	0.005	469,000	dm	0.20	3.13	[33]
Spinach seeds	soil	15,000	dm	0.005	499,000	dm	0.20	3.33	[33]
Corn leaves	water	1,000	fm	0.005	22,000	dm	0.15	3.37	[34]
Lettuce Leaves	soil	5,800	fm	0.030	154,000	dm	0.05	3.50	[31]
Spinach root	soil	15,000	dm	0.005	528,000	dm	0.20	3.52	[33]
Bean leaves	water	5,000	fm		18,600	fm		3.72	[29]
Corn leaves	soil	15,000	dm	0.005	633,000	dm	0.20	4.22	[33]
Radish roots	water	2	fm		9	fm		4.50	[30]
Tomato fruit	water	2	fm		11	fm		5.50	[30]
Spinach shoot	soil	15,000	dm	0.005	832,000	dm	0.20	5.55	[33]
Lettuce Leaves	soil	5,800	fm	0.064	117,750	dm	0.05	5.58	[31]
Lettuce Leaves	soil	5,800	fm	0.067	117,960	dm	0.05	5.88	[31]
Lettuce Leaves	soil	5,800	fm	0.019	405,000	dm	0.05	5.99	[31]
Bean leaves	water	10,000	fm		97,000	fm		9.70	[32]
Bean leaves	water	5,000	fm		96,600	fm		19.32	[29]
Carrot shoot	soil	15,000	dm	0.005	4,294,000	dm	0.20	28.63	[33]

## Figure Legends

Figure 1. Illustration of the pathways by which chemical agents are transferred from soil and soil solution and then into roots, stems, leaves and edible plant tissues.  $K_d$  is the soil solids/soil solution partition coefficient, RCF the root concentration factor, TSCF the transpiration stream concentration factor, SCF the stem concentration factor, and  $K_{ps}$  the generalized plant-to-soil bioconcentration ratio typically relating dry soil solids concentration to dry-mass (edible) vegetation concentrations.

Figure 2. Comparison of the BCR estimates for several models with  $\log K_{ow}$  as the predictive parameter. Also included in this figure are the calibration data for each model and (dark) lines showing our estimate of the four standard deviation interval or 95% confidence interval about the BCR value based on the different models and data.

Figure 3. A cumulative probability plot for the 81 observed values of RDX BCR in Table 2 showing that the cumulative probability of these data based on rank order fits a cumulative lognormal distribution with a geometric mean (GM) BCR of 1.12 and geometric standard deviation (GSD) of 3.5.

Figure 4. Cumulative probability plot showing the relative variance in experimental variability and model uncertainty for estimating the BCR value for a chemical such as RDX that has an expected BCR value of 1.12. Also shown here are estimated 95% confidence intervals on the BCR estimate based on model uncertainty and experimental variability.

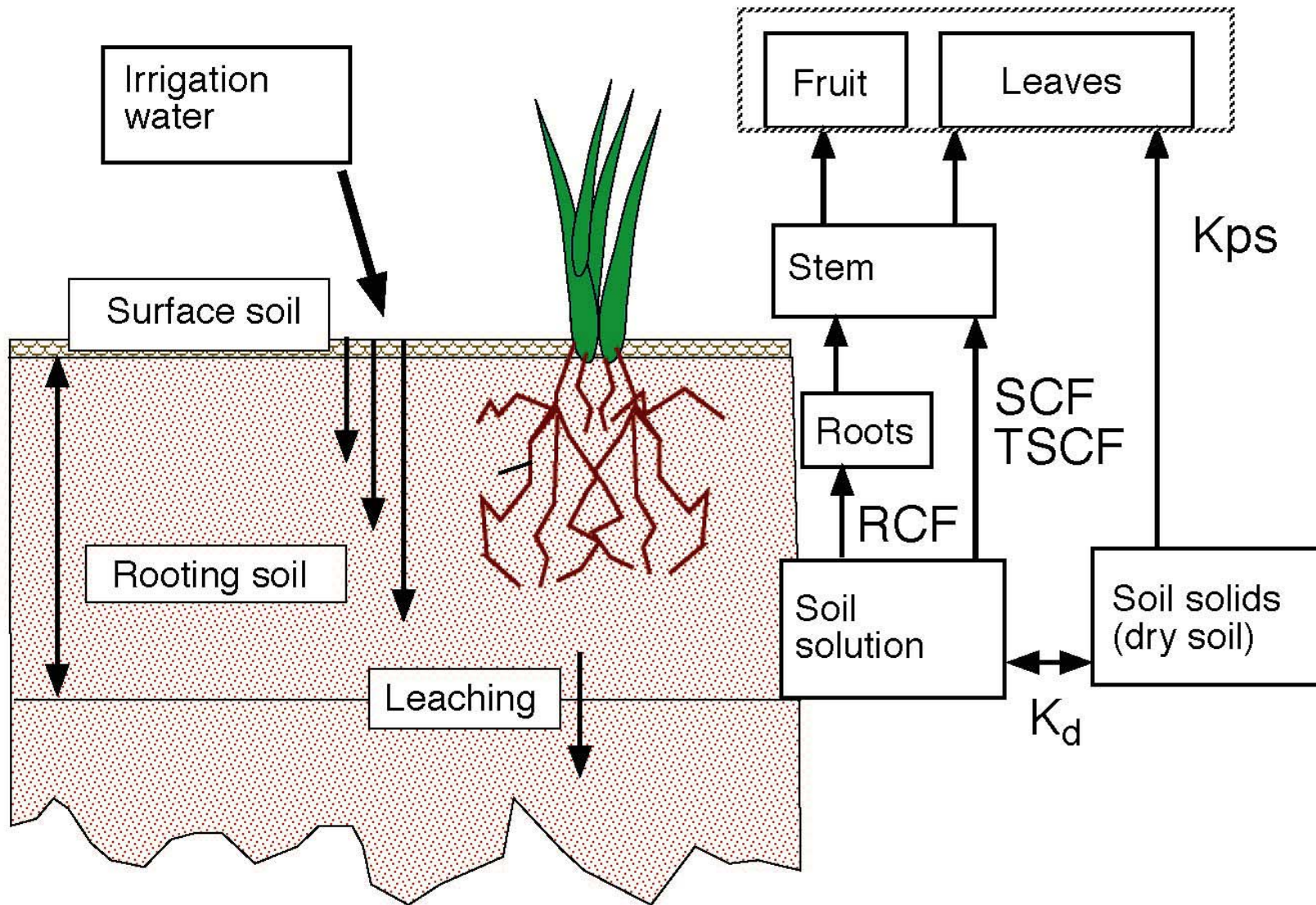


Figure 1

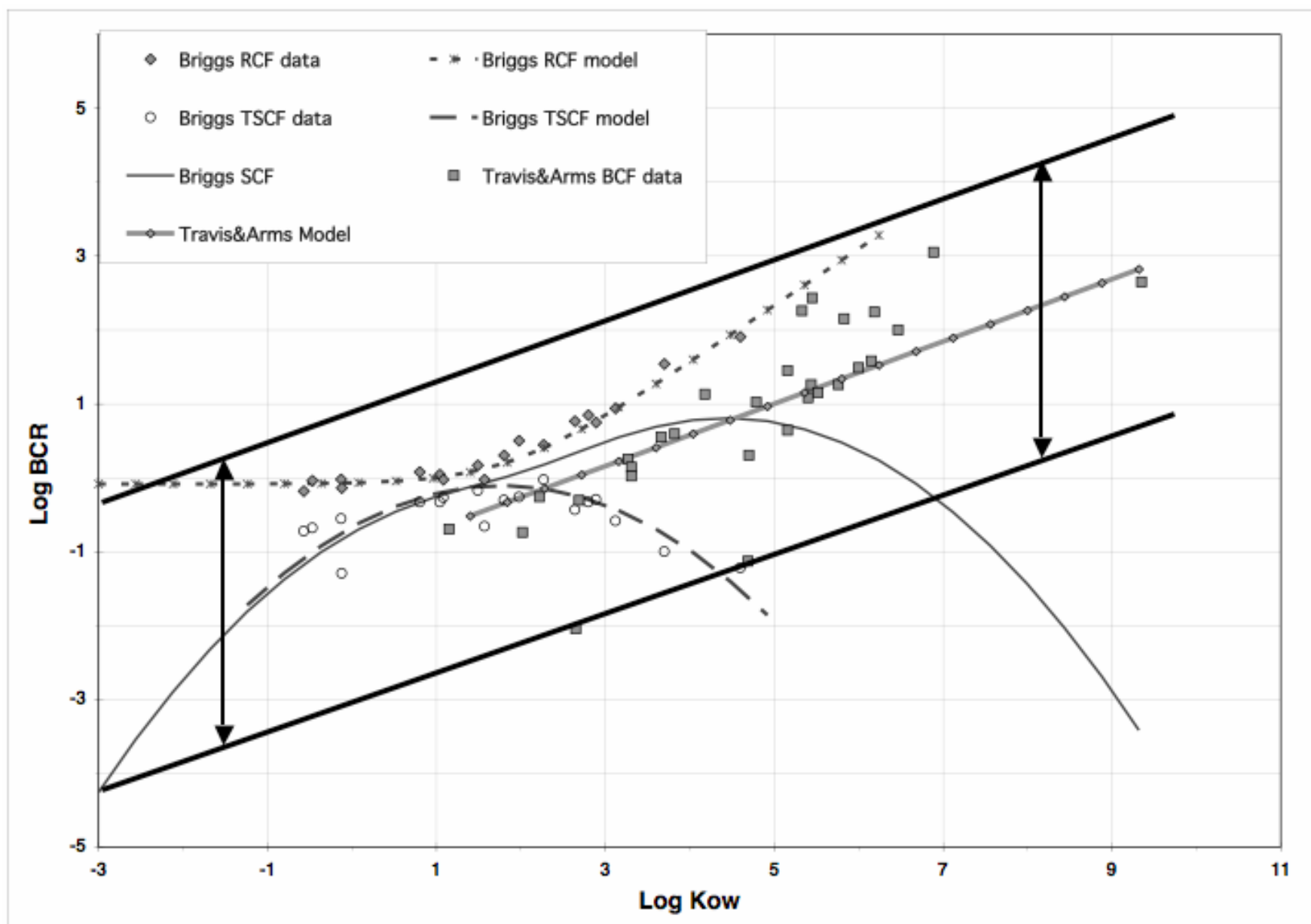


Figure 2

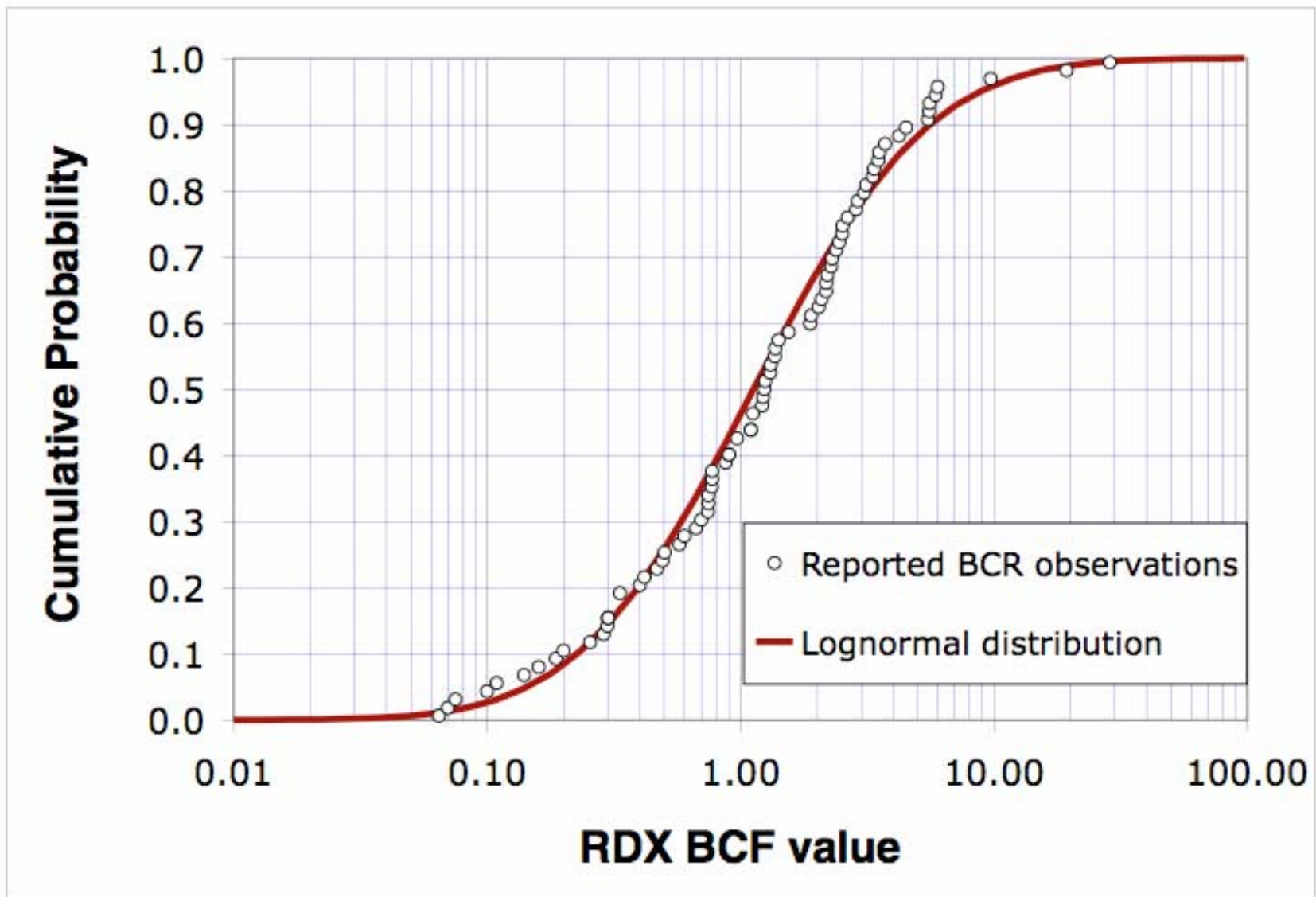


Figure 3

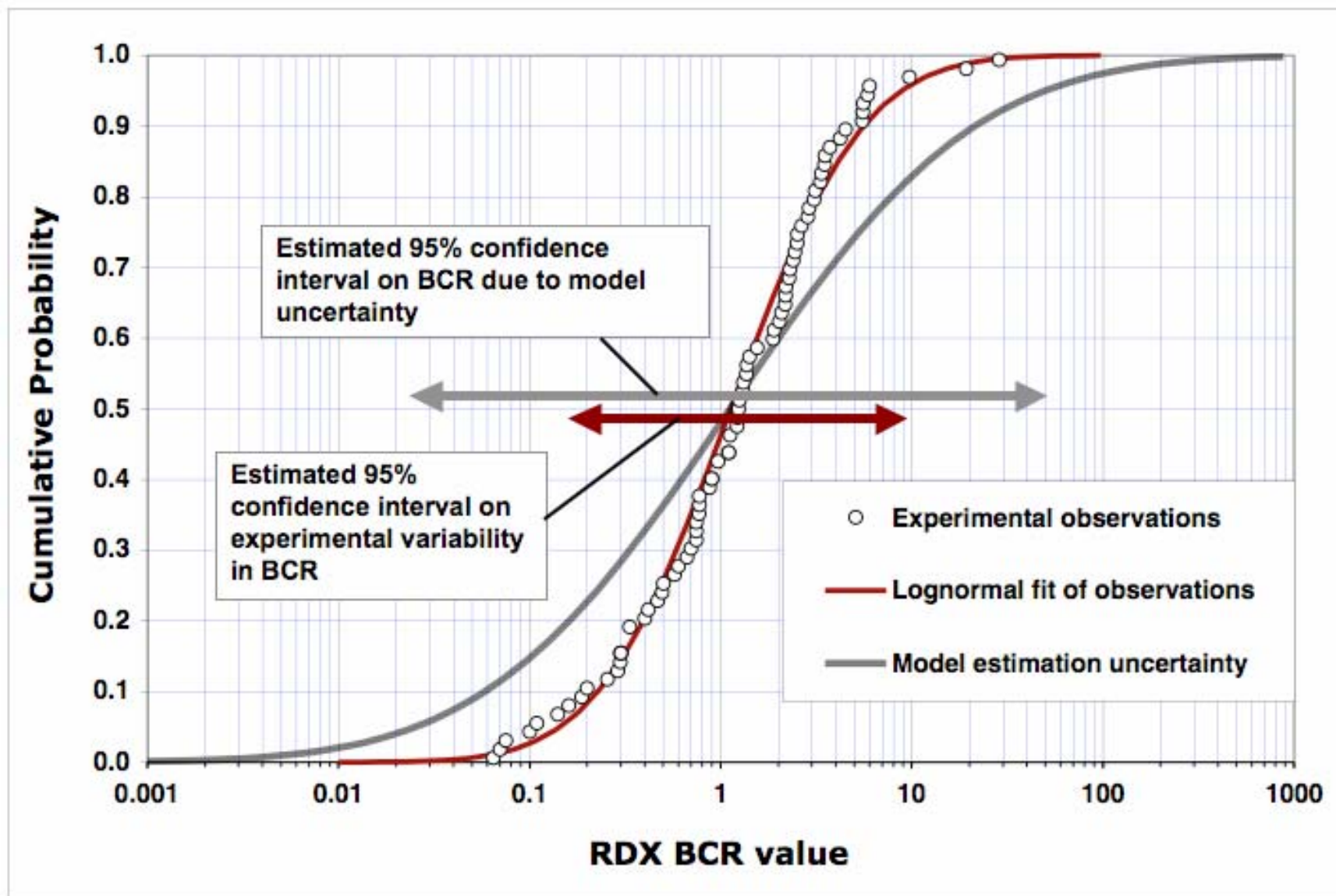


Figure 4